Relation between the Step Pattern and the Velocity of the Moving Linear Adatom Source*

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During deposition of Ga atoms on a Si(111) vicinal face, a step on the vicinal face shows a comb-like pattern. Keeping the formation of comb-like pattern in mind, we carry out phase field simulations, in which a linear adatom source advances in front of a step. The comb-like pattern consisting of straight finger-like intrusions is formed when the source velocity is smaller than a critical value determined by the step anisotropy. Initially, the straight step is unstable and step wandering is induced by the asymmetry of the surface diffusion field. The amplitude of step fluctuations increases with time and an array of short intrusions is formed. Shorter intrusions cannot catch sufficient adatoms, so that coarsening of the pattern occurs. When the adatom source moves slowly, the intrusions grow long and the step shows a regular comb-like pattern. By the coarsening, the distance between intrusions is several times larger than the initial value. The pattern is metastable for a rapid change in the source velocity. When the adatom source moves fast, the intrusions cannot keep up with the adatom source and an irregular pattern is formed. When the strength of crystal anisotropy is weak, splitting of the tips of intrusions frequently occurs and the step shows an irregular seaweed-like pattern. With a strong crystal anisotropy, the step shows a dendrite pattern. [DOI: 10.1380/ejssnt.2015.269]

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I. INTRODUCTION

In many nonequilibrium conditions, step wandering occurs and various step patterns are observed on a vicinal face. For example, the step wandering is caused by an asymmetry of the diffusion filed with the Ehrlich-Schwoebel (ES) effect [1–3]. Taking account of the ES effect, Pierre-Louis et al. [4, 5] studied wandering of steps during growth. They derived a nonlinear evolution equation of the step motion and showed the formation of grooves. The asymmetry of the surface diffusion field may be also due to the drift flow of adatoms. On Si vicinal faces, the drift flow is induced by a direct electric current, and the step wandering is observed during sublimation [6–11]. In the experiments, the step distance is much smaller than the terrace width and the shape of adatoms can be neglected. We studied the step wandering induced by the drift flow without evaporation [12, 13]. The step wandering occurs when the drift is in the downhill direction and the grooves are formed by the in-phase wandering, which agrees with the experiments [6–11].

Behavior of a wandering step in nonconserved systems is different from that in conserved systems. Bena et al. [14] studied the step wandering of an isolated step caused by the ES effect. Carrying out the reductive perturbation analysis, they derived the Kuramoto-Sivashinsky (KS) equation [15, 16] and showed that the motion of an isolated step is chaotic [14, 17]. We carried out a similar analysis for the step wandering induced by the drift flow of adatoms. The wandering pattern is described by the KS equation when the drift flow is perpendicular to the step [18]. With a tilted drift, the pattern becomes regular and is expressed by the Benney equation [19, 20].

Recently, Hibino et al. [21] observed the step motion on a Si(111) vicinal face during Ga deposition. The step shows a comb-like pattern consisting of finger-like intrusions. The distance between neighboring intrusions is much smaller than the terrace width and the shape of intrusions is fairly regular, so that the pattern is different from that in the previous studies [4–13]. The cause to induce the instability is very different from the drift of adatoms and the ES effect. In Ref. [21], the authors suggested that the comb-like pattern is induced by the reconstruction of surface structure. During Ga deposition, the surface structure initially changes from the $7 \times 7$ structure to the $\sqrt{3} \times \sqrt{3}$ structure. Then, the surface structure is transformed from the $\sqrt{3} \times \sqrt{3}$ structure to

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the 6.3 × 6.3 structure. The latter starts from the step edge and the 6.3 × 6.3 structure preferentially spreads into the lower side terrace. During the second surface structural transition, Si atoms are emitted from the solid to the surface at the phase boundary of the two different surface structures. The released Si atoms are supplied to the steps from the lower side terrace and solidification occurs at the steps. The asymmetry of the supply of adatoms probably induces the comb-like step pattern.

The phase boundaries, which act as the source of adatoms, are straighter than the steps. Therefore, in the previous studies [22, 23] we assumed that the source of adatoms moving in front of the step is straight and carried out Monte Carlo simulations by use of a lattice model. We showed that a regular comb-like pattern is formed with a slow adatom source. Due to coarsening, the distance between the final finger-like intrusions is a few times longer than that in the initial stage. In the lattice model [22, 23], controlling some parameters independently such as the crystal anisotropy and the noise is not possible, so that we could not investigate the dependence of final pattern on these parameters.

In this paper we use another model, phase field model, and study the dependence of step patterns on the physical parameters. In Sec. II, we introduce our model [24], which is a phase field model formulated by Karma et al. [25, 26]. In Sec. III, we show results of our simulations. We investigate the change in the step pattern with the strength of the crystal anisotropy and the velocity of the linear source. We also investigate the stability of the comb-like pattern. In Sec. IV, we summarize our results.

II. MODEL

We consider a step along the x-axis advancing to the y-direction. Taking account of the experiment [21], we neglect impingement and evaporation of adatoms. In order to determine the motion of the step, we use a phase filed model, which is similar to that of Ref. [25]. Solving the time evolution equations of the phase field variable ϕ(x, y) and the dimensionless supersaturation of adatom density u, we determine the step motion. The phase field variable ϕ(x, y) represents the height of the surface: ϕ(x, y) = 1 in the upper side terrace and ϕ(x, y) = −1 in the lower side terrace. The variable ϕ(x, y) changes from −1 to 1 in a thin transition region. We regard the location ϕ(x, y) = 0 as the step position.

Time evolution of the phase field variable ϕ is determined by the free energy F, which is given by

\[ F = \int dx dy \left\{ \frac{1}{2} W(\theta)^2 |\nabla \phi|^2 + F \right\}. \]  (1)

The first term is the increase in the free energy density owing to spatial change of the phase field variable. The coefficient W(θ) represents the width of the interface, in which θ is the angle between the normal direction of the step and the y-direction. In our simulation, we assume that W(θ) has a fourfold orientational symmetry: W(θ) = W_0(1 + c_4 \cos 4θ), where W_0 is the interface width and c_4 is the strength of the anisotropy of the step energy. In eq. (1), the second term F is given by

\[ F(\phi, u) = f(\phi) - \lambda u g(\phi), \]  (2)

where f(ϕ) represents a double well potential, whose two minima at the upper side terrace and the lower side terrace are the same as in equilibrium. g(ϕ) represents the difference in the free energy between the solid phase, ϕ = 1, and the vapor phase, ϕ = −1. The parameter λ is a positive constant coupling the phase filed with the density field. The dimensionless supersaturation u is defined as u = Ω(\epsilon(x, y) - c^0_{eq}), where Ω is the atomic area, c(x, y) is the adatom density, and c^0_{eq} is the equilibrium adatom density. In our simulation, f(ϕ) and g(ϕ) are defined as follows:

\[ f(\phi) = -\frac{\phi^2}{2} + \frac{\phi^4}{4}, \]  (3)

\[ g(\phi) = \phi - \frac{2\phi^3}{3} + \frac{\phi^5}{5}. \]  (4)

The phase variable ϕ is not conserved during solidification, and the time evolution of ϕ is given by [25, 26]

\[ \tau(\theta) \frac{\partial \phi}{\partial t} = -\frac{\delta F}{\delta \phi} \]

\[ = \nabla \cdot [W(\theta)^2 \nabla \phi] - \frac{\partial F(\phi, u)}{\partial \phi} \]

\[ + \frac{\partial}{\partial x} \left[ \nabla \phi W(\theta) \frac{\partial W(\theta)}{\partial \phi_x} \right] + \frac{\partial}{\partial y} \left[ \nabla \phi W(\theta) \frac{\partial W(\theta)}{\partial \phi_y} \right], \]  (5)

where \( \phi_x = \partial \phi/\partial x, \phi_y = \partial \phi/\partial y, \) and τ(θ) represents the relaxation time.

In Ref. [26], by taking a thin interface limit, Karma and Rappel showed the relation between a phase field model and a sharp interface model. W(θ) and τ(θ) are related to the kinetic coefficient K(θ) and the capillary length \( \tilde{d}_0(\theta) \), which is expressed as \( \tilde{d}_0(\theta) = \sqrt{2 \epsilon_{eq}(\delta(\theta)/\kappa_B T} \) with the step stiffness \( \delta(\theta) \). The relations between the parameters are given by

\[ K(\theta) = \frac{\lambda W(\theta)}{a_1 \tau(\theta)} \left[ 1 - a_2 \left( \frac{W(\theta)^2}{D \tau(\theta)} \right) \right]^{-1}, \]  (6)

\[ \tilde{d}_0(\theta) = \frac{a_1}{\lambda} \left( W(\theta) + \frac{d^2W(\theta)}{d\theta^2} \right) = d_0(1 - 15\epsilon_4 \cos 4\theta), \]  (7)

where D is the diffusion coefficient, \( d_0 = a_1 W_0/\lambda \) with \( a_1 = 5\sqrt{2}/8 \) and \( a_2 = 0.6267 \). When the step kinetics is infinitely fast, \( \tau(\theta) \) is given by

\[ \tau(\theta) = \frac{a_2 \lambda W(\theta)^2}{D} = \frac{a_2 \lambda W_0^2}{D} (1 + \epsilon_4 \cos 4\theta)^2. \]  (8)

Taking account of the change in ϕ by solidification, the diffusion equation of \( u \) is given by

\[ \frac{\partial u}{\partial t} = D \nabla^2 u - \frac{1}{2} \frac{\partial \phi}{\partial t} - \nabla \cdot q, \]  (9)

where the second term in the right hand side represents the change in the adatom density by solidification. The last term is the change in the adatom density by thermal noise. Since evaporation and impingement of adatoms are neglected, we use a conserved current noise satisfying

\[ \left\{ q_i(r, t) q_j(r', t') \right\} = 2D F_0 \delta_{ij} \delta(r - r') \delta(t - t'). \]  (10)
To reproduce the thermal equilibrium fluctuation, the strength of the noise $F_\nu$ is expressed as $F_\nu = \Omega^2 \langle c_\nu^2 \rangle$ [27]. In our simulation, we study how the step pattern changes with the source velocity and the crystal anisotropy.

III. NUMERICAL SIMULATION

We use a two dimensional square lattice in numerical simulations. The grid width is $a = 1$, and the system size is $L_x \times L_y$. We assume that the linear source and the step advance toward the positive $y$-direction. Initially, a straight step is located at $y_s = 100$: $\phi|_{y \leq y_s} = 1$ and $\phi|_{y > y_s} = -1$. The regions where $y \leq y_s$ and $y \geq y_s$ represent the upper terrace and the lower terrace, respectively. The position of the linear source $y_p$ is initially $y_p = 103$, which is immediately in front of the step. In the $x$-direction, we use the periodic boundary condition. In the $y$-direction, the values of $\phi$ and $u$ satisfy the following conditions, $\phi|_{y=0} = 1$, $\phi|_{y=y_p} = -1$, $\partial u/\partial y|_{y=0} = 0$, and $D \partial^2 u/\partial y^2|_{y=y_p} = -V_p(u|_{y=y_p}-u_0)$, where $V_p$ is the source velocity.

In our numerical simulation, the diffusion coefficient is $D = 1$, the capillary length is $d_0 = 0.05$, and the width of interface is $W_0 = 3$. Taking account of the experiment [21], we use $u_0 = 0.5$ in our simulation. The time increase in a numerical iteration is $\Delta t = 0.2$. We move the source position with the velocity $V_p$: In each time interval $\Delta t/V_p$, we increase $y_p$ by $a$. During simulation, $\phi$ and $u$ are kept constant as $\phi = -1$ and $u = u_0$ in the region where $y_p < y < L_y$. The values of $\phi$ and $u$ in $0 \leq y \leq y_p$ are updated in the simulation.

First, we investigate the change of step pattern with the source velocity. In Fig. 1 we show snapshots with a slow linear source. The solid region where $\phi > 0$ is colored blue, and the position of the linear source is expressed by the red line. Initially, the distance between the linear source and the step is very short and the step is straight. With increasing time, the distance increases and the step starts to fluctuate with a small amplitude. In an early stage [Fig. 1(a)], the step wandering occurs and short intrusions are produced. The gradient of $u$ near the tips of the intrusions is large, so that solidification of adatoms occurs preferentially at the tips and the intrusions grow long [Fig. 1(b)]. Both long intrusions and short ones coexist in the middle stage. Since the long intrusions capture adatoms more efficiently than the short ones, coarsening occurs and only the long intrusions remain. In a later stage [Fig. 1(c)], an array of regular intrusions appears.

In Fig. 2 we show snapshots in which the linear source moves faster than that in Fig. 1. The step patterns in Figs. 2(a) and 2(b) are similar to Figs. 1(a) and (b). In the late stage [Fig. 2(c)], however, splitting of tips frequently occurs and the pattern is more irregular than Fig. 1(c). The source velocity in Fig. 3 is faster than that in Fig. 2. Like the irregular pattern in Fig. 2, the step shows an irregular seaweed-like pattern. However, the tips of intrusions cannot catch up with the linear source, while the tips move at the same velocity as the linear source in Fig. 2.

From the time evolution of the tip velocity, we can find when the intrusions start to grow rapidly. Figure 4 shows the time evolution of the ratio of the fastest tip velocity $V_s$ to the source velocity $V_p$. In the case that $V_p = 5 \times 10^{-3}$, the ratio $V_s/V_p$ exceeds unity when $t = 4.7 \times 10^4$. In Fig. 1, intrusions become evident at the time. The intrusions advance fast by preferential solidification at the tip positions, so that the distance between tips and the source starts to decrease. In the case of $V_p = 5 \times 10^{-2}$, the maximum of the distance appears earlier than in $V_p = 5 \times 10^{-3}$ but the overshoot of the ratio $V_s/V_p$ is smaller, so that the converged distance is slightly larger. In the case of $V_p = 1.5 \times 10^{-2}$, the tip velocity never exceeds the source velocity and the distance between the highest tip and the source increases monotonically.

In the case of $V_p = 5 \times 10^{-3}$ coarsening finishes and regular intrusions grow steadily when $t \geq 2.0 \times 10^5$. To confirm the end of coarsening, we observe the time evolution of the period of intrusions [Fig. 5]. We define the period of intrusions as the system width $L_x$ divided by the number of intrusions. In an early stage, the period of intrusions is smaller than 100. When $t \approx 6.0 \times 10^4$, the period starts to increase rapidly. Then, the increasing rate becomes gradually slow. The period is saturated when $t > 1.5 \times 10^5$. Thus, the time evolution of the period...
of intrusions is consistent with the result in Fig. 4.

Next, we investigate how the step pattern depends on the crystal anisotropy $\epsilon_4$. Figure 6 shows the morphology diagram. The step patterns are classified into four types. When the tips of intrusions follow the linear source, the step pattern shows a regular comb-like pattern or an irregular seaweed-like pattern. With increasing $\epsilon_4$, the parameter region with regular pattern increases. The parameter $\epsilon_4$ stabilizes the comb-like pattern. When the tips do not follow the linear source, the step pattern is a dendrite pattern with side intrusions or irregular seaweed-like pattern. The irregular seaweed-like pattern is formed when the crystal anisotropy $\epsilon_4$ is small. With a large $\epsilon_4$, splitting of the tip is suppressed, but the side intrusions are formed and the step shows a dendrite pattern [Fig. 7]. In the morphology diagram, the solid line below which the step follows the adatom source can be determined precisely by observing the distance between the linear source and the tip position of the highest intrusion. However, the positions of the other boundaries indicated by dotted lines are obscure because we determine the boundaries by looking at the step pattern.

Finally, we focus on the regular comb-like pattern and its period. Figure 8 shows the relation between the intrusion period in a late stage, $\Lambda$, and the source velocity $V_p$. When $V_p$ is sufficiently small ($V_p \leq 0.01$), the period $\langle \Lambda \rangle$ averaged over the samples becomes shorter with increasing $V_p$ as $\langle \Lambda \rangle \sim V_p^{-1/2}$. This relation agrees with the experiment [21], the lattice model [22,23,28] and our previous study [24]. When $V_p$ is large ($V_p > 0.01$), the period $\langle \Lambda \rangle$ becomes longer with increasing $V_p$. Increasing of $\langle \Lambda \rangle$ is not seen in the lattice model [22, 23]. In the phase field model the step has a finite width, so that the structure finer than the step width $W(\theta)$ is not formed. Thus, DLA(diffusion-limited-aggregation)-like pattern seen in our lattice model, does not appear and the comb-like pattern of a large period is formed. In Ref. [29], Brener et al. studied the motion of a needle shaped crystal in a narrow channel of supercooled melt. They obtained a relation between the channel width and the velocity of the needle shaped crystal, which is similar to the relation between $\langle \Lambda \rangle$ and $V_p$ in Fig. 8. It was
pointed out that the finger-like interface moving slowly is unstable. In their model, the temperature is kept constant far from the interface and the crystal grows freely. The straight linear source is present in front of the step in our model, so that fluctuations of the tips of intrusions are suppressed. The existence of the linear source in front of the step is very likely to stabilize the comb-like pattern.

In order to see the stability of the comb-like pattern, we investigate how the regular pattern is affected by a rapid change in the source velocity. Figure 9 shows the time evolution of $\langle \Lambda \rangle$. First, we make a steady comb-like pattern, in which $\langle \Lambda \rangle$ and $V_P$ are given by the point A in Fig. 8. Then, we suddenly change $V_P$ from $5 \times 10^{-3}$ to $8 \times 10^{-3}$. The step follows the adatom source and the state is rapidly transformed from the point A to the point B. However, the transition from the point B to the point C is stable.

![FIG. 4. Ratio of the fastest tip velocity to $V_P$. The crystal anisotropy is $\epsilon_4 = 10^{-2}$, and the noise strength is $F_u = 10^{-5}$. The data are averaged over 10 runs. The error bars are smaller than the marks.](image1)

![FIG. 5. Time evolution of the period between intrusions. The source velocity is $V_P = 5 \times 10^{-3}$. The crystal anisotropy is $\epsilon_4 = 10^{-2}$, and the noise strength is $F_u = 10^{-5}$. The data are averaged over 10 runs.](image2)

![FIG. 6. Morphology diagram. In orange, purple, blue, and green regions, a comb-like pattern, an irregular seaweed-like pattern following the adatom source, an irregular seaweed-like pattern not following the adatom source, and a dendrite pattern are formed, respectively. In the upper side of a solid line, the step cannot follow the linear adatom source.](image3)

![FIG. 7. A snapshot of an irregular dendrite pattern in a late stage. The velocity of the linear source is $V_P = 0.018$. The crystal anisotropy is $\epsilon_4 = 5 \times 10^{-2}$, and the noise strength is $F_u = 10^{-5}$.](image4)

![FIG. 8. Dependence of the period $\langle \Lambda \rangle$ between intrusions in a late stage on $V_P$. Solid circles show $\langle \Lambda \rangle$ obtained by a straight step growing in a large system. The crystal anisotropy is $\epsilon_4 = 10^{-2}$, and the noise strength is $F_u = 10^{-5}$. The data are averaged over 10 runs. Open circles show the velocity $V_P$ of a free needle crystal growing in a channel system, in which the channel width is $\Lambda$ and $F_u = 0$.](image5)

![FIG. 9. Time evolution of $\langle \Lambda \rangle$ with a sudden change in $V_P$. If a straight step grows with $V_P = 5 \times 10^{-3}$ and $8 \times 10^{-3}$, $\langle \Lambda \rangle$ converges to the value given by the upper and the lower dotted lines, respectively. After a stable comb-like pattern is formed with $V_P = 5 \times 10^{-3}$, $V_P$ is changed to $8 \times 10^{-3}$. The arrow shows the time at which $V_P$ is changed.](image6)
point C, which is the steady state realized by the growth of a straight step with $V_p = 8 \times 10^{-3}$, does not occur. The decrease in $\Lambda$ is not induced by the rapid change in the source velocity and the period is kept unchanged. Also, the decrease in the source velocity from $8 \times 10^{-3}$ to $5 \times 10^{-3}$ does not induce an increase in $\Lambda$. Thus, the comb-like pattern seems metastable for a small change in $V_p$.

IV. SUMMARY

In this paper, keeping the comb-like pattern formed on a Si(111) vicinal face in mind [21], we studied the wandering of a step guided by the linear adatom source. Since the adatom source is present in the lower side of step, the surface diffusion field is asymmetric and the step wandering occurs.

In the phase filed model [24], the anisotropy of the step energy and the noise are controlled easily than those in the lattice model [22,23,28]. Using the model, we clarified the parameter region of $\epsilon$ and $V_p$ where the regular comb-like pattern is formed. In Ref. [24], we mainly studied properties of the regular pattern during steady growth. In this paper, we studied the coarsening process which leads to the steady growth. Initially, a fluctuation with the wavelength expected by the linear stability analysis grows and intrusions are formed. Small fluctuation of the height of the intrusions causes coarsening. From Figs. 4 and 5, it is seen that the coarsening stops when the survived intrusions catch up with the adatom source. This is probably because the fluctuation of the tip height is suppressed.

In our previous study by the lattice model [23], we showed that the period of the regular pattern changes with sudden change in the source velocity. In this paper, we investigated the stability of regular pattern for a small change in source velocity. The period between intrusions remains unchanged with the small velocity change, so that the comb-like pattern looks like metastable for a small velocity change.

If the source of adatoms is sufficiently far from the step, our system looks similar to the growth model in a supercooled melt [30–32]. In the melt growth model, the interface patterns are classified in more detail than those in our system. We intend to study the relation between our model and the growth from the supercooled melt in the future.

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